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STEW: A Nonlinear Data Modeling Computer Program

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Abstract

A nonlinear data modeling computer program, STEW, employing the Levenberg-Marquardt algorithm, has been developed to model the experimental ^{239}Pu(n,f) and ^{235}U(n,f) cross sections. This report presents results of the modeling of the ^{239}Pu(n,f) and ^{235}U(n,f) cross-section data. The calculation of the fission transmission coefficient is based on the double-humped-fission-barrier model of Bjornholm and Lynn. Incident neutron energies of up to 5 MeV are considered.

1 Introduction

The ^{239}Pu(n,f) and ^{235}U(n,f) cross sections have been well measured[1, 2]. Figure 1 shows the latest ENDL evaluation of the ^{239}Pu(n,f) and ^{235}U(n,f) cross sections up to 5 MeV of incident neutron energy. The evaluation of ^{239}Pu(n,f) is based on an analysis of 33 sets of experimental data whereas the evaluation of ^{235}U(n,f) is based on an analysis of 60 sets of data. The estimated uncertainty of the evaluation is less than 2%[3]. To calculate the fission cross section, the double-humped fission barrier model of Bjornholm and Lynn[4] is usually employed. Calculated cross sections from this model can be made to best-fit the experimental data, in the least-square sense, by adjusting the parameters in the model. The consistency of the parameters obtained from various fits can be used to determine the applicability of the physical model.

When conservation of angular momentum and parity is enforced in the simulation of reaction dynamics, a physical model of nuclear fission is necessary despite the existence of experimental fission cross sections. This can be seen from the Hauser-Feshbach[5] formulation of the fission cross section calculation:

$$\sigma_{\mathrm{n,f}}(E_i, I, P) = \sum_{J,\pi} \sigma_{\mathrm{R}}(E_i, I, P; U, J, \pi) \frac{\Gamma_{\mathrm{f}}(U, J, \pi)}{\Gamma(U, J, \pi)} \tag{1}$$

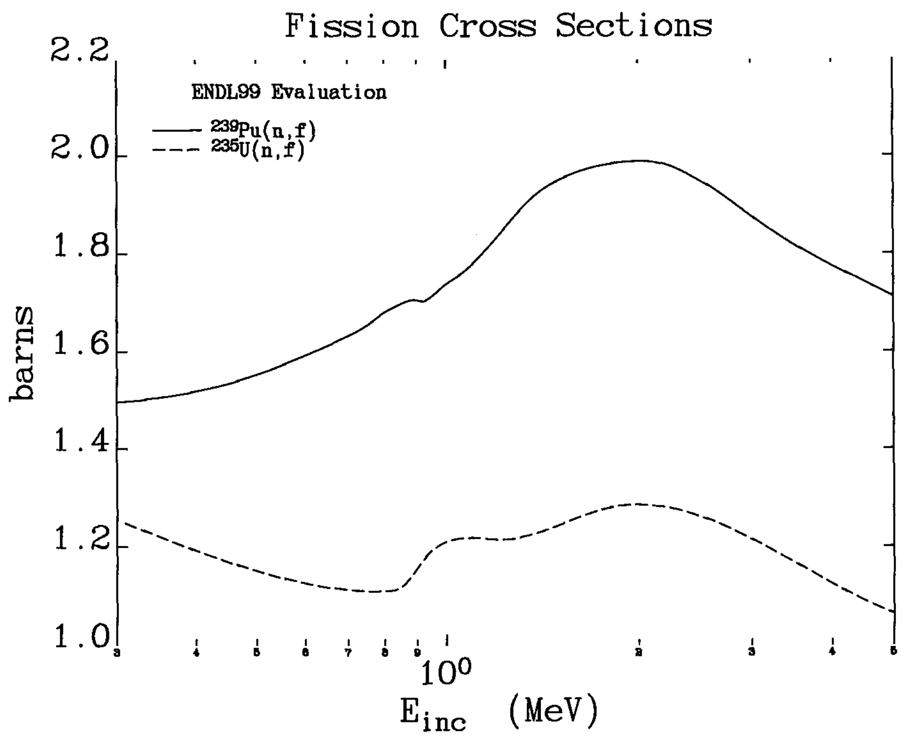

Figure 1: *Experimental evaluation of ^{239}Pu(n,f) and ^{235}U(n,f) cross sections as a function of incident neutron energy.*

where the left-hand-side is the cross section of neutron-induced fission on a target with spin I and parity P. The center of mass energy of the neutron-and-target system is denoted by E_i. The excitation energy, angular momentum, and parity of the compound nucleus are U, J, π, respectively. The reaction cross section, $\sigma_{\mathrm{R}}(E_i, I, P; U, J, \pi)$, describes the probability of the compound-nucleus formation. The fission and total decay widths, $\Gamma_{\mathrm{f}}(U, J, \pi)$ and $\Gamma(U, J, \pi)$, can be related to transmission coefficients through the reciprocity theorem[6], and $\Gamma(U, J, \pi)$ is a sum of widths of all possible decay channels. In order to treat fission as one of the competing channels of the compound reaction, the fission width $\Gamma_{\mathrm{f}}(U, J, \pi)$ as a function of U, J, π is required and must be provided by a physical model.

Calculations of various cross sections of the neutron induced reactions on fissile isotopes, such as ^{239}Pu and ^{235}U, show that fission cross section is the dominant component of the reaction cross section[7, 8]. A small change in fission cross section can have a large effect on some of the weaker channels such as (n,n') or (n,2n), for example. It is therefore important to constrain the calculated fission cross section to the known experimental values in order to reduce the uncertainty in the calculated cross sections of the non-fission channels. Such need was heightened

	V_A	$\hbar\omega_A$	V_B	$\hbar\omega_B$
^{240}Pu	5.57	1.04	5.07	0.6
^{236}U	5.63	1.04	5.53	0.6
	C_{A1}	θ_{A1}	C_{B1}	θ_{B1}
^{240}Pu	0.015	0.284	0.271	0.343
^{236}U	0.015	0.284	0.027	0.343
	C_{A2}	θ_{A2}	C_{B2}	θ_{B2}
^{240}Pu	1.6	0.4265	0.5	0.5
^{236}U	1.6	0.5	0.4265	0.5

Table 1: *Initial parameters used in calculating fission cross sections for the ^{240}Pu and ^{236}U nuclei.*

in our modeling of the n+^{239}Pu and n+^{235}U reactions. The computer program, STEW, has therefore been written in order to accomplish the task of constraining the parameters in the fission model such that the calculated fission cross sections best fit the experimental data.

STEW is a stand-alone computer program. The computer program, chosen by the user, that carries out the model calculations is called by STEW. The user determines which parameters in the model calculations are to be adjusted. These parameters in the input file are marked by a marker selected also by the user. This stand-alone feature of STEW leaves the model-calculation component completely arbitrary and therefore allows it to be applied to the modeling of any physical quantity for which there exists experimental data.

2 The Fission Model Used

By adding shell corrections to the Liquid Drop Model[9], Strutinsky[10] pioneered the work that lead to the double-humped fission barrier model of Bjornholm and Lynn[4]. We use this model to simulate the nuclear fission process. In the model, the shapes of the two fission barriers are approximated by two parabolas. The transmission coefficient of a nucleus, with excitation energy E, through a single barrier is given by Hill and Wheeler[11] as:

$$T(J,\pi,E) = \int_0^\infty \frac{\hat{\rho}(J,\pi,\epsilon)d\epsilon}{1+\exp\left[V+\epsilon-E\right]/\hbar\omega} \tag{2}$$

where V is the fission barrier height and $\hbar\omega$ is the curvature of the barrier at the saddle point. The dependence of the level density $\hat{\rho}(J,\pi,E)$ on parity is taken to be a constant:

$$\hat{\rho}(J,\pi=-1,E) = \hat{\rho}(J,\pi=+1,E) = \frac{1}{2}\tilde{\rho}(J,E)\,. \tag{3}$$

The functional dependence of $\tilde{\rho}(J,E)$ on angular momentum, J, is given by a Gaussian:

$$\tilde{\rho}(J,E) = \rho_J(J)\rho(E) = \frac{(2J+1)\exp\left[-(J+1/2)^2/2\sigma^2\right]}{2\sqrt{2\pi}\sigma^2}\rho(E)\,, \tag{4}$$

and the dependence of $\rho(E)$ on the excitation energy, E, is assumed to be of the constant temperature form:

$$\rho(E) = \sum_J \tilde{\rho}(J,E) = C\exp E/\theta \tag{5}$$

where θ is the nuclear temperature and C is a constant. The spin-cutoff parameter, σ^2, which characterizes the Gaussian in J, is assumed to be independent of the excitation energy. The two fission barriers, labeled by A and B, are assumed to be uncorrelated so that the total transmission coefficient through both barriers is simply:

$$T(J,\pi,E) = \frac{T_A(J,\pi,E)T_B(J,\pi,E)}{T_A(J,\pi,E)+T_B(J,\pi,E)}\,. \tag{6}$$

Once the fission transmission coefficient is obtained through this model, it is used to calculate the fission cross section by employing the statistical description of nuclear reactions given by Hauser and Feshbach[5].

Following the suggestions of Bjornholm and Lynn[4], the excitation energy, ϵ (c.f. Equation (2)), which is measured from the top of the barriers, is divided into two ranges. They are, in units of MeV: $0 \leq \epsilon < 3$ and $3 \leq \epsilon$. Initial values, which are again based on suggestions from Bjornholm and Lynn, of the two fission barrier heights, their corresponding curvatures and level density parameters, are given in Table 1. Since ϵ is divided into two ranges, two sets of level density parameters are used. The level density parameters (c.f. Equation (5)) in the first range of excitation energy above barriers A and B are labeled by $C_{A1}, \theta_{A1}, C_{B1}, \theta_{B1}$ whereas those in the second range of excitation energy are labeled by $C_{A2}, \theta_{A2}, C_{B2}, \theta_{B2}$. These are the parameters that are adjusted in order to best fit the experimental fission cross sections. For incident neutron energy of up to 5 MeV, only first-chance fission needs to be considered.

3 The Optimization Algorithm Used

The process of adjusting parameters in a theoretical model to obtain the best fit to experimental data is a problem of minimization in a multi-dimensional space[12, 13]. Since the functional dependence of the fission transmission coefficient on the parameters in Table 1 is not linear, we are faced with a nonlinear optimization problem which is difficult in general. Several methods which address the issue of nonlinear data modeling are described in reference [13]. We adopted the Levenberg-Marquardt method[14], which provides an algorithm for varying smoothly between the inverse-Hessian method and the steepest decent method[13].

Consider a set of M unknown parameters $\vec{a} \equiv \{a_k; k = 1, 2, ..., M\}$ in a model described by

$$y = y(x; \vec{a}) \tag{7}$$

where x is the independent variable. The best-fit parameters are those that give rise to the minimum of the χ^2 merit-function defined by

$$\chi^2(\vec{a}) = \sum_{i=1}^{N} \left[\frac{y_i - y(x_i; \vec{a})}{\sigma_i} \right]^2 \tag{8}$$

where y_i is the experimental value at x_i and σ_i is the standard deviation of the data point (x_i, y_i). The total number of points is N. When the χ^2 is near its minimum, the inverse-Hessian method is used. Let $\vec{a}_{min}$ be the value of $\vec{a}$ at which χ^2 is a minimum and $\vec{a}_{cur}$ be the current value of $\vec{a}$. The inverse-Hessian method gives the solution of $\delta\vec{a} = \vec{a}_{min} - \vec{a}_{cur}$ by

$$\sum_{l=1}^{M} \alpha_{kl} \delta a_l = \beta_k \tag{9}$$

where

$$\alpha_{kl} \equiv \frac{1}{2} \frac{\partial^2 \chi^2}{\partial a_k \partial a_l} = \sum_{i=1}^{N} \frac{1}{\sigma_i^2} \left[\frac{\partial y(x_i; \vec{a})}{\partial a_k} \frac{\partial y(x_i; \vec{a})}{\partial a_l} - [y_i - y(x_i, \vec{a})] \frac{\partial^2 y(x_i; \vec{a})}{\partial a_l \partial a_k} \right] \tag{10}$$

$$\beta_k \equiv -\frac{1}{2} \frac{\partial \chi^2}{\partial a_k} = \sum_{i=1}^{N} \frac{y_i - y(x_i; \vec{a})}{\sigma_i^2} \frac{\partial y(x_i; \vec{a})}{\partial a_k} . \tag{11}$$

When $\chi^2(\vec{a})$ is far from $\chi^2(\vec{a}_{min})$, the steepest decent method is used which gives a solution of $\delta\vec{a}$ by

$$\delta a_k = constant \times \beta_k . \tag{12}$$

Recognizing that the magnitude of the constant in Equation (12) must be proportional to $1/\alpha_{kk}$ and combining Equations (9) and (12), Marquardt arrived at the equation:

$$\sum_{i=1}^{M} \alpha'_{kl} \delta a_l = \beta_k \tag{13}$$

where

$$\alpha'_{kk} \equiv \alpha_{kk}(1 + \lambda) \tag{14}$$

$$\alpha'_{kl} \equiv \alpha_{kl} \qquad (k \neq l) \tag{15}$$

and λ is a dimensionless parameter. The variation between Equations (9) and (12) is achieved by adjusting the value of λ. When λ is large, the α' matrix becomes diagonally dominant and Equation (13) approaches Equation (12). On the other hand, when λ becomes very small, Equation (13) approaches Equation (9). By giving an initial guess for the set of M parameters, $\vec{a}$, and an initial value of λ, one can iteratively solve for $\delta\vec{a}$ from Equation (13) by adjusting the value of λ until a preset condition for terminating the iteration is satisfied. A detailed recipe, which we followed, of such a procedure is given on page 679 of reference [13]. The termination condition we use is the following:

- *if* $\chi^2 \leq 0.01$ *or the fractional change of* χ^2 *is less than or equal to* 0.1%.

In calculating the elements of the α matrix, the second-derivative term in Equation (10) is ignored for reasons of stability[13]. The first derivatives with respect to the parameters $\vec{a}$ are calculated numerically which leaves the functional dependence of $y(x;\vec{a})$ on $\vec{a}$ completely arbitrary.

4 Results and Discussions

Based on the algorithm outlined in Section 3, the computer program STEW has been written to search for the parameters given in Table 1 in order to obtain the best fit to the evaluated fission cross sections shown in Figure 1.

The model calculation includes two reaction mechanisms: the direct and the compound. The direct reaction mechanism is described by the optical model[15]. The compound reaction mechanism is described by the statistical reaction model of Hauser-Feshbach[5], and the fission process is considered as one of the competing channels of the compound reaction. As both the ^{239}Pu and ^{235}U nuclei are deformed, the coupled-channel option in the optical model is employed and the theoretical tool used for this component of the calculation is the ECIS code[16]. The first 5 discrete states of the ground state band of the ^{239}Pu and ^{235}U nuclei

	V_A	$\hbar\omega_A$	V_B	$\hbar\omega_B$
^{240}Pu	5.71	1.07	5.05	1.68
^{236}U	5.61	1.03	5.53	0.64
	C_{A1}	θ_{A1}	C_{B1}	θ_{B1}
^{240}Pu	0.015	0.177	0.039	0.433
^{236}U	0.012	0.272	0.892	0.694
	C_{A2}	θ_{A2}	C_{B2}	θ_{B2}
^{240}Pu	0.00001	0.1501	0.025	0.4201
^{236}U	0.066	0.3776	0.201	0.4717

Table 2: *Final parameters obtained from the optimization program STEW for the* ^{240}*Pu and* ^{236}*U nuclei.*

are coupled. We use the phenomenological optical model potential of Dietrich[17]. The optical-model parameters given by Dietrich are used in the fission-parameter search by STEW for the ^{235}U nucleus. For the ^{239}Pu nucleus, the optical-model parameters from Dietrich are used as the initial values. The optical model parameter search routine BIGLAZY[18, 19] is then used to slightly modify these parameters to give the best fit to the experimental total cross section, ^{239}Pu(n,tot), and to the available angular distribution data for elastic scattering[1]. Ground-state deformation parameters used for the ^{239}Pu nucleus are: $\beta_2 = 0.2$ and $\beta_4 = 0.06$ as they give rise to the best BIGLAZY fit. For the ^{235}U nucleus, these deformation parameters are taken from the calculations by Möller and Nix[21] as: $\beta_2 = 0.22$ and $\beta_4 = 0.08$. The Hauser-Feshbach component of various reaction channels is calculated using the reaction modeling code POLIFEMO which is part of the IDA[18] system of codes. In the energy range of up to 5 MeV of incident neutron energies, the compound reaction channels include capture, compound elastic scattering, inelastic scattering and fission. STEW computes and iteratively minimizes the $\chi^2(\vec{a})$ by taking the experimental data and repeatedly calling POLIFEMO. The initial values of fission parameters, that is, the initial values of $\vec{a}$, are given in Table 1. Table 2 shows the results of the STEW optimization.

In order to reduce the runtime of the optimization procedure, the number of data points in the ENDL99 evaluation for both ^{239}Pu(n,f) and ^{235}U(n,f) is reduced. This data reduction is assisted by the computer program THINNER[20] which preserves the shape of the evaluated (n,f) curve while reducing the number of data points. A standard deviation of 2% of the evaluated fission cross section is assigned to each data point for both ^{239}Pu(n,f) and ^{235}U(n,f) evaluations. The total number of parameters adjusted is 12. However, not all parameters are allowed to vary simultaneously at the beginning. Typically, we let 4 parameters to be adjusted at one time. The best-fit values are then taken as constants when another 4 parameters are allowed to vary. Once all 12 parameters have been separately adjusted, we let all 12 parameters to simultaneously vary and obtain the final best-fit values. Results shown in Table 2 are from the final iteration. In general, the optimization process is time-consuming. We run STEW on a Sun workstation employing four 300 MHz UltraSPARC-II processors (only one processor is used at one time). With the ECIS calculations carried out beforehand and results stored, the runtime for the minimization ranges from about 20 minutes to a few hours. The length of time depends on the number of parameters that are being simultaneously adjusted, the number of iterations required to obtain the best fit, as well as other details of the minimization procedure, among which are the stopping condition and the step size of the λ parameter in the Levenberg-Marquardt algorithm.

Figures 2 and 3 show comparisons of the calculated fission cross sections and the experimental evaluations. The calculations are performed using parameters in Table 1 and those in Table 2, separately. The curve corresponding to the set of fission parameters in Table 1 is labeled by "Bjornholm + Lynn parameters"

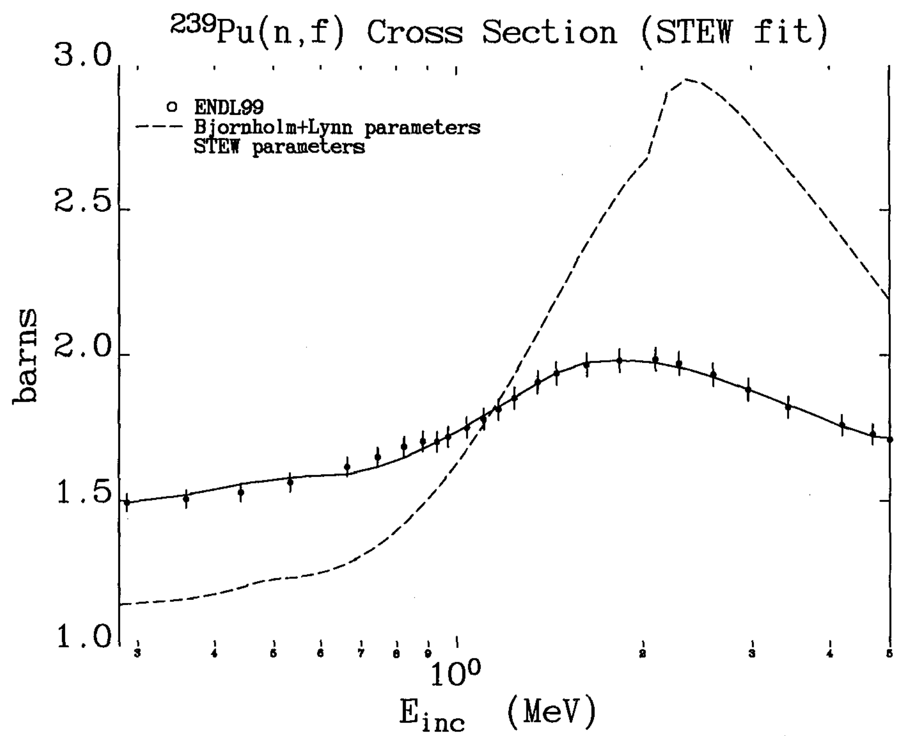

Figure 2: *A STEW fit of the ^{239}Pu(n,f) cross section, based on the double-humped-fission-barrier model of Bjornholm and Lynn, as a function of incident neutron energy.*

whereas the curve corresponding to the set of parameters in Table 2 is labeled by "STEW parameters". One sees, from these figures, that the parameters are well constrained in that the agreement between the model calculation and experimental data is much improved.

A close examination of Tables 1 and 2 shows that the change of parameters as a result of the optimization process mostly occurs to those that are used in the constant-temperature level density given by Equation 5. The change in fission barrier heights is less than 150 keV in all cases, consistent with the uncertainties from the experimental determination of the barrier heights[22]. With the exception of $\hbar\omega_B$ for the ^{240}Pu nucleus, the change in curvatures is also very small, ranging from 0.6% to 7%. The increase of $\hbar\omega_B$ in ^{240}Pu from 0.6 MeV to 1.68 MeV means that in order to get a good fit to the experimental data, the outer barrier, that is, barrier B, of ^{240}Pu must be narrower which leads to a higher penetration probability. On the other hand, the changes in the level-density parameters are rather dramatic. The most noticeable characteristic of the change in these parameters is not so much in the absolute magnitude of the values, but in the

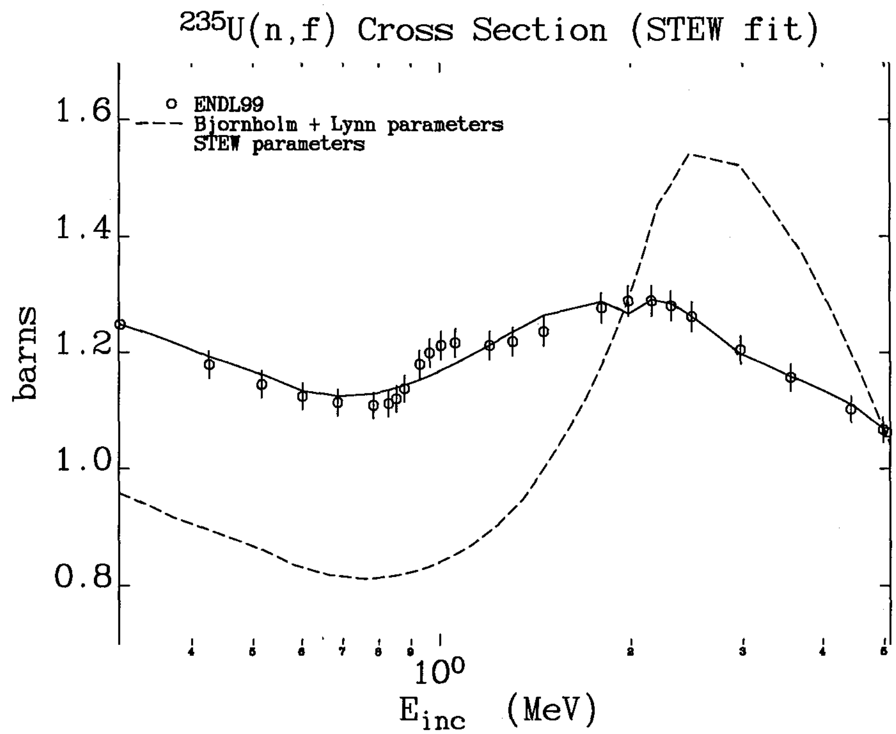

Figure 3: *A STEW fit of $^{235}U(n,f)$ cross section, based on the double-humped-fission-barrier model of Bjornholm and Lynn, as a function of incident neutron energy.*

relative magnitude of the nuclear temperature. The nuclear temperature should increase with the excitation energy of the nucleus, which means that $\theta_{A2,B2}$ should be greater than $\theta_{A1,B1}$. Table 2 shows that this is not the case except for the level density above the first barrier in ^{240}U. Since STEW tries to best minimize the χ^2 of Equation 8 for the pre-selected model of Equation 7, this unphysical feature in the optimized level-density parameters suggests a possible inadequacy of the constant-temperature-level-density model in describing the nuclear fission process. One piece of physics that is absent in the constant-temperature level density is the collective enhancement due to the large deformation of the nucleus before fission[6]. Such collective effect is most pronounced when the difference between the excitation energy of the nucleus and the fission barrier height is not large. However, it should be pointed out that the nuclear structure in a pre-scission nucleus is not known well enough to uniquely determine the correct level density formulation. This introduces an element of arbitrariness in the choice of level density and the associated correction factors in the calculation of fission cross sections. Another missing element in the fission model used is the damping in the second well which describes the interaction of the fission motion with the internal degrees of freedom[23]. The absence of such a description could also have contributed to the unphysical behavior of the level density parameters observed in Table 2. We also note that while the continuity of the experimental data ensures the overall continuity of the calculated fission cross sections during the optimization process, the explicit continuity in the constant-temperature level density between the two ranges of excitation energy is not enforced. This gives rise to the small dip in the modeled ^{235}U(n,f) cross section at about 2 MeV of incident neutron energy. At this incident energy, the excitation energy of the ^{236}U nucleus is in the region where the chosen first and second excitation-energy ranges meet.

5 Conclusions

To conclude, STEW successfully optimized the fission parameters in the incident energy range of up to 5 MeV. The generality of the program allows it to be applied to any type of nonlinear data modeling. Also revealed in the optimization process is the possible deficiencies of the constant-temperature-level-density model used in calculating fission cross sections. Thus, by examining the physical content of the best-fitted parameters, the data modeling procedure presented in this report provides a means of investigating the soundness of the physical models employed in the calculation of a given physical quantity.

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